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14. ABSTRACT This research project studies complex multielectron effects in isolated atoms and molecules. The study helps explore the overlap between atomic and condensed matter physics, particularly the usage of atomic physics to learn about many-body phenomena. Multielectron effects constitute many-body problems in atomic systems. However, the multielectron problems have not been traditionally appreciated in AMO physics since most problems in atoms and molecules can be understood through the single-independent-electron picture. Multielectron effects, however, become noticeable when atoms or molecules are exposed to an intense electromagnetic field. In this project, the PI's team has made significant progresses in studying multielectron correlation effects in atoms and molecules in strong laser fields. These results allow us gain further understanding in many-body concepts, which may shine light to many correlated electron problems in condensed materials that are strategically important to Air Force Scientific Research. Tangentially, we have recently created a femtosecond laser processing technique to transform highly reflective metals to a nearly perfect light absorber or only selectively absorbing a certain color of light.						
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Final Report to AFOSR

PROJECT TITLE

How Much Can We Learn About Many-Body Concepts From A
Single Atom or Molecule?

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Abstract

This research project studies complex multielectron effects in isolated atoms and molecules. The study helps explore the overlap between atomic and condensed matter physics, particularly the usage of atomic physics to learn about many-body phenomena in condensed materials. Multielectron effects constitute many-body problems in atomic systems. However, the multielectron problems have not been traditionally appreciated in AMO physics since most problems in atoms and molecules can be understood through the single-independent-electron picture. Multielectron effects, however, become noticeable when atoms or molecules are exposed to an intense electromagnetic field.

In this project, the PI's team has made significant progresses in studying multielectron correlation effects in atoms and molecules in strong laser fields. The study spans correlated electron phenomena that involve one-, two-, and three-electron dynamics. These results allow us gain further understanding in many-body concepts, which may shine light to many correlated electron problems in condensed materials that are strategically important to Air Force Scientific Research.

Tangentially, we have recently created a femtosecond laser processing technique to transform highly reflective metals to a nearly perfect light absorber or only selectively absorbing a certain color of light, so the metals appear either pitch black or having a certain distinct color. The darkened and colored metals have many important applications such as making better sensors, detectors, solar energy collectors, laser marking, and improved stealth technology. This work has garnered extensive public interests recently and was covered by public and scientific media reports such as The New York Times, MSNBC, Discovery Channel News, Science, and Nature Magazines.

Project Results

In this project, we have made significant progresses in studying multielectron correlation effects in atoms and molecules in strong laser fields. The study spans on studying correlated electron phenomena that involve one-, two-, and three-electron dynamics. Additionally, we have also created a femtosecond laser processing technique to transform highly reflective metals to a nearly perfect light absorber or only selectively absorbing a certain color of light. Some of our results with publications are summarized below.

Published Research Results

Atoms and Molecules:

1. Wavelength effects on strong-field single electron ionization
J. Wu and C. Guo, Adv. Studies in Theo. Phys. **2**, 271 (2008).
2. Polarization effects on nonsequential double ionization of molecular fragments in strong laser fields, J. Wu, H. Zeng, and C. Guo, Phys. Rev. A **75**, 043402 (2007).
3. Single-ionization-induced dissociation of heteronuclear diatomic molecules in strong fields, J. Wu, H. Zeng, and C. Guo, J. Phys. B **40**, 1095 (2007).
4. Multielectron effects of diatomic molecules in strong laser fields (Book Chapter),
C. Guo, in "*Progress in Ultrafast Intense Laser Science*", vol. **1**, 43 (Springer Series in Chemical Physics, 2006).
5. Comparison study of atomic and molecular single ionization in the multiphoton ionization regime, J. Wu, H. Zeng, and C. Guo, Phys. Rev. Lett. **96**, 243002 (2006).
6. Triple-ionization-induced dissociation of NO in strong laser fields
J. Wu, H. Zeng, and C. Guo, Phys. Rev. A (Rapid Comm.) **74**, 031404(R) (2006).
7. Holding molecular dications together in strong laser fields
C. Guo, Phys. Rev. A (Rapid Comm.) **73**, 041401(R) (2006).
8. Dynamics of triple ionization induced dissociation in diatomic molecules in strong fields, J. Wu, H. Zeng, J. Wang, and C. Guo, Phys. Rev. A (Rapid Comm.) **73**, 051402(R) (2006).
9. Vertical and nonvertical transitions in triple-ionization-induced dissociation of diatomic molecules, J. Wu, H. Zeng, and C. Guo, Phys. Rev. A **74**, 065403 (2006)
10. Slowing down molecular dissociation in strong laser fields
C. Guo, *Ultrafast Phenomena XV* (Springer-Verlag, 2006).
11. Non-sequential double ionization in slow charge fragmentation of doubly ionized NO
J. Wu, H. Zeng, and C. Guo, J. Phys. B **39**, 3849 (2006).
12. Attosecond pulse XUV photoionization in two color and chirped laser field,
Y. Huo, Z. Zeng, R. Li, Z. Xu, C. Guo, Z. Sun, Y. Rhee, Opt. Lett. **30**, 564 (2005).
13. Observation of anomalous charge separation following single electron strong field ionization, C. Guo, Phys. Rev. A (Rapid Comm.) **71**, 021405(R) (2005).

14. Channel competition between metastable and dissociated states of doubly ionized NO in strong laser fields, C. Guo and K. Wright, Phys. Rev. A (Rapid Comm.) **71**, 021404(R) (2005).
15. Ellipticity effects on nonsequential double ionization in diatomic molecules, C. Guo, J. Phys. B (*Letters to the Editor*) **38**, L323 (2005). **(Featured article of the issue)**

Metals:

16. Spectral and polarization responses of femtosecond laser-induced periodic surface structures on metals
A. Y. Vorobyev and C. Guo, J. Appl. Phys. **103**, 043513 (2008);
17. Colorizing metals with femtosecond laser pulses
A. Y. Vorobyev and C. Guo, *Optics & Photonics News* **19**(12), 30 (2008).
18. Femtosecond laser-induced periodic surface structure formation on tungsten
A. Y. Vorobyev and C. Guo, J. Appl. Phys. **104**, 063523 (2008).
19. Femtosecond laser blackening of platinum
A. Y. Vorobyev and C. Guo, J. Appl. Phys. **104**, 053516 (2008).
20. Modeling of residual thermal effect in femtosecond laser ablation of metals: role of a gas environment, N.M. Bulgakova, V.P. Zhukov, A.Y. Vorobyev and C. Guo, Appl. Phys. A **92**, 883 (2008).
21. Colorizing metals with femtosecond laser pulses
A. Y. Vorobyev and C. Guo, Appl. Phys. Lett. **92**, 041914 (2008).
(Selected into March 2008 issue of Virtual Journal of Ultrafast Science).
22. Dissipative nanostructures and Feigenbaum's universality in the "metal-high-power ultrashort-pulsed polarized radiation" nonequilibrium nonlinear dynamical system
V.S. Makin, R.S. Makin, A.Y. Vorobyev, and C. Guo, Tech. Phys. Lett. **34**, 387 (2008).
23. Extraordinary enhanced absorptivity of gold surface ablated with femtosecond laser pulses" A. Y. Vorobyev, C. Guo, V. S. Makin, N. G. Kokody, and V. M. Kuzmichev , Proc. SPIE **7009**, 700913 (2008) .
24. Resolving dynamics of acoustic phonons by surface plasmons
J. Wang, J. Wu, and C. Guo, Opt. Lett. **32**, 719 (2007).
25. Periodic ordering of random surface nanostructures induced by femtosecond laser pulses on metals, A.Y. Vorobyev, V.S. Makin, and C. Guo, J. Appl. Phys. **101**, 034903 (2007).
26. Femtosecond laser structuring of titanium implants
A.Y. Vorobyev and C. Guo, Appl. Surf. Sci. **253**, 7272 (2007).
27. Effects of nanostructure-covered femtosecond laser-induced periodic surface structures on optical absorptance of metals
A.Y. Vorobyev and C. Guo, Appl. Phys. A (Rapid Comm.) **86**, 321 (2007).
28. Residual thermal effects in laser ablation of metals
A.Y. Vorobyev and C. Guo , J. Phys. CS **59**, 418 (2007).
29. Effect of electron heating on femtosecond laser-induced coherent acoustic phonons in noble metals, Phys. Rev. B **75**, 184304 (2007).
30. Change in absorptance of metals following multi-pulse femtosecond laser ablation
A.Y. Vorobyev and C. Guo , J. Phys. CS **59**, 579 (2007).

31. Shot-to-shot correlation of residual energy and optical absorptance in fs laser ablation
A.Y. Vorobyev and C. Guo, App. Phys. A, **86**, 235 (2007).
32. Permanent recording of light helicity on optically-inactive metal surfaces
J. Wang and C. Guo, Opt. Lett., **31**, 3641 (2006).
33. Enhanced energy coupling in femtosecond laser-metal interactions at high intensities
A.Y. Vorobyev and C. Guo, Opt. Express **14**, 13113 (2006).
34. Observation of a step change in optical absorption of gold in vacuum
Z. Ma and C. Guo, Phys. Rev. B **74**, 233404 (2006).
35. Femtosecond laser nanostructuring of metals
A.Y. Vorobyev and C. Guo, Opt. Express **14**, 2164 (2006).
36. Formation of extraordinarily uniform periodic structures on metals induced by fs laser pulses
J. Wang and C. Guo, J. Appl. Phys. **100**, 023511 (2006).
37. Thermal effects in femtosecond laser ablation of metals
C. Guo (**Invited paper**), Proc. SPIE **6118**, 611808 (2006).
38. Residual thermal effects in Al following ns- and fs-laser pulse ablation
A.Y. Vorobyev, V.M. Kuzmichev, N.G. Kokody, P. Kohns, J. Dai, and C. Guo, App. Phys. A, **82**, 357 (2006).
39. Mechanism of surface structural modification on absorptivity of metals in multi-pulse femtosecond laser ablation, A.Y. Vorobyev, C. Guo, V.S. Makin, N.G. Kokody, V.M. Kuzmichev, Proc. SPIE (2006).
40. Ultrafast dynamics of femtosecond laser-induced surface pattern formation on metals
J. Wang and C. Guo, App. Phys. Lett. **87**, 251914 (2005).
41. Direct observation of enhanced residual thermal energy coupling to solids in femtosecond laser ablation
A.Y. Vorobyev and C. Guo, App. Phys. Lett. **86**, 011916 (2005).

Some selected summaries on Research Results (Mainly in Atomic and Molecular Physics):

1. Wavelength effects on strong-field single electron ionization
J. Wu and C. Guo, Adv. Studies in Theo. Phys. **2**, 271 (2008).

In this paper, we perform *ab initio* quantum mechanical calculations on wavelength dependence of strong-field single electron ionization on two model atoms with different initial electron distributions. The numerical results show dramatically different wavelength dependence for different initial electron distributions.

2. Comparison study of atomic and molecular single ionization in the multiphoton ionization regime
J. Wu, H. Zeng, and C. Guo, Phys. Rev. Lett. **96**, 243002 (2006).
(Selected into the July 2006 issue of Virtual Journal of Ultrafast Science).

In this paper, we report, for the first time in the multiphoton ionization regime, a comparison study of single-electron ionization of diatomic molecules versus rare gas

atoms with virtually the same ionization potentials. In comparing N_2^+ to Ar^+ , a higher ion signal is seen in N_2^+ compared to Ar^+ for linear polarization but the difference vanishes in circularly polarized light. In comparing O_2^+ to Xe^+ , we observe a suppression in O_2^+ compared to Xe^+ for both linear and circular polarization but this suppression exhibits an intensity dependence, i.e., there is little suppression for O_2^+ at the lowest intensity range, but the suppression becomes increasingly stronger as the laser intensity increases. The multielectron screening model is used to discuss possible mechanisms of this intensity dependent suppression in O_2^+ in the multiphoton ionization regime.

3. Polarization effects on nonsequential double ionization of molecular fragments in strong laser fields

J. Wu, H. Zeng, and C. Guo, *Phys. Rev. A* **75**, 043402 (2007).

(Also selected into the May 2007 issue of Virtual Journal of Ultrafast Science).

In this paper, we study non-sequential double ionization in various molecular fragments for laser polarization aligning either parallel or perpendicular along the molecular axis. For the first time in diatomic molecule O_2 , we find that non-sequential double ionization can be turned on and off when we simply switch the laser polarization between perpendicular ionization and parallel ionization. Furthermore, we find that the dependence of kinetic energy release on intensity can be used to distinguish sequential versus non-sequential ionization in molecules. The results in this paper allow us to gain better understanding on the role of non-sequential ionization in molecular dissociative ionization.

4. Single-ionization-induced dissociation of heteronuclear diatomic molecules in strong fields

J. Wu, H. Zeng, and C. Guo, *J. Phys. B* **40**, 1095 (2007).

In this paper, we introduce a general technique to study dissociation channels of singly ionized heteronuclear diatomic molecules by comparing the branching ratios of these channels induced by a light field either parallel or perpendicular to the molecular axis. The experimental results show that a singly ionized heteronuclear diatomic molecule, if it dissociates, will be strongly influenced by its detailed electronic structure. Our study of single-ionization-induced dissociation demonstrates the fundamental difference between ionization dynamics in strong fields versus weak fields.

5. Resolving dynamics of acoustic phonons by surface plasmons

J. Wang, J. Wu, and C. Guo, *Opt. Lett.* **32**, 719 (2007).

(Selected into the March 2007 issue of Virtual Journal of Ultrafast Science).

In this paper, we show that the dynamics of acoustic phonons generated by femtosecond impulsive optical excitation can be clearly resolved by a surface plasmon technique, with an enhanced sensitivity orders of magnitude higher than regular optical probe measurements. Our calculations confirm that the enhanced sensitivity is an intrinsic property of the surface plasmon probe. Therefore, the surface plasmon

technique is a promising tool to detect small signal changes in optical and mechanical properties on a microscopic scale.

6. Slowing down molecular dissociation in strong laser fields

C. Guo, in *Ultrafast Phenomena XV* p564 (Springer-Verlag, New York, 2007).

Doubly ionized carbon monoxide always appeared to dissociate in previous strong-field experiments and the metastable channel was scantily seen. In this paper, we demonstrate experimental conditions to obtain an abundance of metastable CO^{2+} .

7. Super-sensitive Surface Plasmon Probe in Ultrafast Measurements

J. Wang and C. Guo, *Optics & Photonics News* **18**(12), 37 (2007).

Time-resolved femtosecond (fs) pump-probe measurements are a powerful method for studying electron and lattice dynamics in solid materials. In this work, we show that the dynamics of acoustic phonons generated by femtosecond impulsive optical excitation can be clearly resolved by a surface plasmon technique, with an enhanced sensitivity orders of magnitude higher than regular optical probe measurements.

8. Looking at femtosecond laser-induced black metals at different polarizations

A. Y. Vorobyev and C. Guo, *Optics & Photonics News* **18**(12), 43 (2007).

For the first time, we find that optical absorptance of metals can be significantly enhanced by a new type of surface structures following femtosecond laser ablation, namely nanostructure-covered periodic surface structures. Especially, the effect of the nanostructure-covered periodic structures on optical absorptance of metals has a clear polarization dependence that suggests a more controllable way to modify material optical properties with femtosecond laser processing.

9. Triple-ionization-induced dissociation of NO in strong laser fields

J. Wu, H. Zeng, and C. Guo, *Phys. Rev. A (Rapid Comm.)* **74**, 031404(R) (2006).
(Selected into the October 2006 issue of Virtual Journal of Ultrafast Science)

In this paper, we study the dynamics of triple-ionization-induced dissociation in a heteronuclear diatomic molecule NO. Compared to homonuclear diatomic molecules, NO shows a greater complexity in its final states following triple ionization. By utilizing a well-established technique from the study of sequential versus non-sequential ionization, our study shows that both the $\text{N}^{2+} + \text{O}^+$ and $\text{N}^+ + \text{O}^{2+}$ channels are predominately formed non-vertically through a relatively slowly dissociating $\text{N}^+ + \text{O}^+$ state. Finally, we show that both the $\text{N}^{2+} + \text{O}^+$ and $\text{N}^+ + \text{O}^{2+}$ channels are formed at nearly the same internuclear separation that is much smaller than the critical internuclear distance, indicating that dissociative ionization of high charge states in a molecule can occur as a molecule steadily expands from its equilibrium separation rather than always at the critical internuclear distance.

10. Permanent recording of light helicity on optically-inactive metal surfaces

J. Wang and C. Guo, *Opt. Lett.*, **31**, 3641 (2006).

(Selected into the December 2006 issue of Virtual Journal of Ultrafast Science).

In this paper, we report on an unusual permanent recording of light helicity on optically achiral metals. Following a number of circularly polarized (CP) or elliptically polarized (EP) femtosecond laser pulses, well-defined periodic surface structures are found on metal surfaces. These surface structures show different orientation when formed by left CP/EP compared to right CP/EP light. The formation of these structures is attributed to the interference between the incident light and the excited surface plasmons. To our knowledge, this is the only phenomenon that can permanently record light helicity with an optically inactive material.

11. Dynamics of triple ionization induced dissociation in diatomic molecules in strong fields

J. Wu, H. Zeng, J. Wang, and C. Guo, *Phys. Rev. A (Rapid Comm.)* **73**, 051402(R)

(2006). (Selected into the June 2006 issue of Virtual Journal of Ultrafast Science)

In this paper, we report the first comparison study between N_2 and O_2 on their triple ionization induced dissociation channels using near-IR 800-nm ultrashort laser pulses. Our experiment shows that the $O^{2+} + O^+$ channel is predominately formed non-vertically through the intermediate $O^+ + O^+$ channel, while the $N^{2+} + N^+$ channel appears to be formed vertically at the lower intensity range but non-vertically at higher intensities. Interestingly, $N^{2+} + N^+$ appears to be in the electronically excited state when it is reached vertically in the lower intensity range. The different triple ionization and dissociation behaviors between N_2 and O_2 can be understood by considering the ionization dynamics of the two molecules in our ultrashort laser pulses.

12. Vertical and nonvertical transitions in triple-ionization-induced dissociation of diatomic molecules

J. Wu, H. Zeng, and C. Guo, *Phys. Rev. A* **74**, 065403 (2006)

(Selected into the January 2007 issue of Virtual Journal of Ultrafast Science)

In this paper, we study triple-ionization-induced dissociation in N_2 and O_2 for laser polarization aligning either parallel or perpendicular along the molecular axis. For the first time, we demonstrate that vertical/nonvertical transition can be turned on and off by simply switching the laser polarization between being perpendicular and parallel to the molecular axis. We also show that the dependence of kinetic energy release on intensity can be used to distinguish vertical from nonvertical transition in triple-ionization-induced dissociation.

13. Enhanced energy coupling in femtosecond laser-metal interactions at high intensities

A.Y. Vorobyev and C. Guo, *Opt. Express* **14**, 13113 (2006).

In this paper, we report on various nanostructures produced through direct surface modification on metals using femtosecond laser pulses. We show, for the first time, that these nanostructures are natural consequence following femtosecond laser ablation. The optimal conditions for producing various nanostructures are determined.

14. Holding molecular dications together in strong laser fields

C. Guo, Phys. Rev. A (Rapid Comm.) **73**, 041401(R) (2006).

(Selected into the May 2006 issue of Virtual Journal of Ultrafast Science).

Metastable channel of doubly ionized carbon monoxide, CO^{2+} , was scantily seen in previous strong-field experiments at visible wavelength region, but was commonly observed using single high-energy-photon/electron excitation. In this paper, for the first time with near-IR ultrashort-pulse radiation, we observe an abundance of CO^{2+} . We show that CO^{2+} results from nonsequential double ionization while its dissociation counterpart, $\text{C}^+ + \text{O}^+$, results from sequential processes, and CO^{2+} can be obtained through either a single high-energy photon/electron excitation or multiphoton ionization with ultrashort pulses before a critical internuclear distance is reached. Our study demonstrates the experimental conditions to converge the outcomes from two vastly different regimes, namely multiphoton excitation and ionization in strong fields and single high-energy-photon/electron excitation and ionization in weak fields.

15. Non-sequential double ionization in slow charge fragmentation of doubly ionized NO

J. Wu, H. Zeng, and C. Guo, J. Phys. B **39**, 3849 (2006).

With ultrahigh time-resolution time-of-flight measurements, we identify a new $\text{N}^+ + \text{O}^+$ dissociation channel following doubly ionized NO. This dissociation channel has a relatively small kinetic energy release. A relatively high non-sequential double ionization rate is found existing in this $\text{N}^+ + \text{O}^+$ channel for linear polarization, but non-sequential double ionization nearly vanishes for circular polarization. These results corroborate the understanding that electronic structures play a key role in influencing non-sequential double ionization in molecules.

Main Personnel involved in this project:

Graduate students: Jincheng Wang and Jian Wu

Postdoctoral Associate: Anatoliy Vorobyev